

## PATENT ABSTRACTS OF JAPAN

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 TECHNOL  
 (22)Date of filing : 11.05.1993 (72)Inventor : KUNIOKA MASAO

## (54) NEW BIODEGRADABLE HIGH WATER ABSORBENT AND ITS PRODUCTION

## (57)Abstract:

**PURPOSE:** To obtain a biodegradable water absorbent, excellent in biocompatibility and useful as a soil improving agent, a material, etc., for artificial skin by dissolving a specific amount of poly( $\gamma$ -glutamic acid) in water, etc., exposing the resultant solution to radiation and then separating the produced cross-linked material.

**CONSTITUTION:** The objective water absorbent having 40-90% gelatinization ratio is obtained by dissolving poly( $\gamma$ -glutamic acid) produced by a microorganism in water or a mixed solvent of the water with a water-soluble solvent such as methyl alcohol so as to provide 1.5-6wt.% concentration thereof, exposing the resultant solution to radiation such as  $\gamma$ -rays preferably at 1.0-50Mrad and then separating the produced cross-linked material.

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CLAIMS

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## [Claim(s)]

[Claim 1] The biodegradability quantity water absorption object which consists of the Pori (gamma-glutamic acid) radiation-induced crosslinking object of 40 - 90% of rates of gelation.

[Claim 2] The manufacture approach of the biodegradability quantity water absorption object characterized by separating the generated bridge formation object after irradiating a radiation at the solution which dissolved Pori (gamma-glutamic acid) in the mixed solvent of water or water, and a water-soluble solvent so that the concentration might become 1.5 - 6% of the weight, and was subsequently obtained.

[Claim 3] The approach according to claim 2 the exposure of a radiation is what irradiates a gamma ray by quantity of radiation 1.0 - 50Mrad.

[Claim 4] The approach according to claim 3 by which Pori (gamma-glutamic acid) is produced by the microorganism

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DETAILED DESCRIPTION

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## [Detailed Description of the Invention]

## [0001]

[Industrial Application] This invention relates to a new biodegradability quantity water absorption object and its manufacture approach. Since the biodegradability quantity water absorption object of this invention receives biodegradation in a nature, it can be used for the modifier in the engineering-works field, the soil conditioner in the plantation art field, a seed coating agent, the water retention agent for vegetable cultivation, the disposable diaper in makeup and the toiletries field, sanitary items, a mechano-chemical material, etc., for example.

## [0002]

[Description of the Prior Art] It is known by using the molecule bridge formation by repeating graft polymerization, radiation-induced crosslinking, ion bridge formation, bridge formation by the cross linking agent, freezing, and fusion to a water soluble polymer etc. that a high water absorption object will generate. These compounds are used abundantly after absorbing a water sordes at the product which can be held so that it may not be leaked, for example, a disposable diaper, sanitary items, etc., using the property of unlike water absorbents, such as cloth and cotton, not repelling water even if it puts a pressure.

[0003] There are the starch system quantity water absorption object which carries out starch as such a water absorption object, carries out the graft polymerization of the synthetic monomers, such as an acrylic acid, to this as a raw material, and is acquired by forming three-dimension bridge formation, a hyaluronic acid system quantity water absorption object which is the water gel of a biological system, a polyvinyl alcohol system quantity water absorption object by which chemosynthesis was carried out, an acrylate system quantity water absorption object, an acrylamide system quantity water absorption object, etc.

[0004] These high water absorption objects will remain without being decomposed into the nature after use, when it is used in a nature like a soil conditioner. Moreover, it is very difficult to collect these. Then, development of the high water absorption object which has biodegradability which after use is decomposed also from on environmental preservation by the microorganism which lives in a nature, and is incorporated by the carbon cycle cycle of a nature serves as pressing need.

## [0005]

[Problem(s) to be Solved by the Invention] This invention conquers the fault which such a conventional high water absorption object has, and is made for the purpose of offering the high water absorption object which can aim at promotion of use in a nature and which has biodegradability.

## [0006]

[Means for Solving the Problem] this invention persons came to make this invention for having the absorptivity in which the bridge formation object of the rate of predetermined gelation obtained by irradiating the radiation of predetermined quantity of radiation by the water solution or aquosity solution of predetermined concentration of Pori (gamma-glutamic acid) which is the water soluble polymer of biodegradability was excellent, without losing biodegradability based on a header and this knowledge, as a result of repeating research variously, in order to develop the

high water absorption object of biodegradability.

[0007] That is, this invention offers the biodegradability quantity water absorption object which consists of the Pori (gamma-glutamic acid) radiation-induced crosslinking object of 40 - 90% of rates of gelation.

[0008] In this invention, the rate of gelation means the percentage of the dry weight of the Pori (gamma-glutamic acid) radiation-induced crosslinking object to the amount of preparation Pori (gamma-glutamic acid).

[0009] The biodegradability quantity water absorption object of this invention dissolves Pori (gamma-glutamic acid) in the mixed solvent of water or water, and a water-soluble solvent so that the concentration may become 2 - 5% of the weight preferably 1.5 to 6% of the weight, and after it irradiates a radiation at the solution subsequently obtained, it can manufacture it by separating the generated bridge formation object.

[0010] There is especially no limit about Pori (gamma-glutamic acid) used in this invention. Although based on what kind of manufacturing methods, such as a manufacturing method, a chemosynthesis method, etc. using a microorganism Thing [for example, Biosci.BioTech.BioChem., produced by microorganisms, such as what produced by the microorganism, for example, a genus *Bacillus* kind like *Bacillus subtilis* etc., 56, 1031-1035 (1992), JP,1-174397,A reference] is desirable and what has 100 [hundreds of thousands -] and hundreds of thousands of molecular weight especially is desirable.

[0011] In this invention, although there is especially no limit about the water-soluble solvent used for dissolving Pori (gamma-glutamic acid) as a mixed solvent with water, it is good to make the rate of water into sufficient amount and to adjust the rate of a water-soluble solvent suitably so that Pori (gamma-glutamic acid) may be dissolved and it may get. As this water-soluble solvent, methyl alcohol, ethyl alcohol, an acetone, methyl acetate, ethyl acetate, etc. are mentioned, for example.

[0012] As for the solution which dissolved Pori (gamma-glutamic acid), it is desirable to put in, seal and use for a radiolucency container, for example, a glass vial bottle etc. Although there is especially no limit about the radiation irradiated by this solution, for example, alpha rays, beta rays, a gamma ray, an electron ray, a neutron beam, an X-ray, a charged-particle line, etc. are mentioned, it makes it possible to use a gamma ray preferably, and to obtain the specified substance by the shape of a film, since beta rays, an electron ray, and alpha rays have small penetrating power.

[0013] the case where a gamma ray is used for such radiation irradiation processing -- advantageous -- a gamma ray -- quantity of radiation, 1.0 to 50 Mrad, preferably, it irradiates by 2 - 30Mrad more preferably, and bridge formation usually advances at a room temperature 1.5 to 40 Mrad.

[0014] Although there is especially no limit about a gamma ray, the thing which made it generate with the irradiation equipment which makes the cobalt 60 a line source, for example is used. In bridge-formation-izing, especially temperature is not important and bridge formation usually advances at a room temperature.

[0015] thus, the obtained rough product -- an aquosity medium -- by carrying out purification processing with water, such as distilled water, preferably, the biodegradability quantity water absorption object which removes a polymer and a decomposition product non-constructed a bridge and consists of the bridge formation object of a desired high grade can be prepared. This purification processing is usually performed by a dipping, dialysis, etc.

[0016] Thus, although the acquired biodegradability quantity water absorption object is transparent and colorless gel, is excellent in absorptivity and is swollen also in the mixed solvent of water and alcohol, or the mixed solvent of an acetone and water, it is not swollen in methyl alcohol, ethyl alcohol, an acetone, a tetrahydrofuran, chloroform, PENZEN, n-hexane, and n-heptane.

[0017] The property of the biodegradability quantity water absorption object of this invention changes with the amount of radiation irradiation, or concentration of said solution, and a degree of cross linking serves as high water absorption small, and by the high exposure and high concentration, although bridge formation will be in a saturation state and serves as low water

absorption relatively, which hold absorptivity sufficient even in those at a low exposure or low concentration.

[0018] The biodegradability quantity water absorption object of this invention can be used repeatedly. Even if it makes it dry by freeze-drying the bridge formation object which absorbed water and makes water swell again, it is changeless to absorptivity ability. Even if it repeated this several times, change was hardly seen by absorptivity ability.

[0019] Moreover, absorptivity ability changes with pH, and the biodegradability quantity water absorption object of this invention has low water absorption in acid pH field, and is high in alkaline pH field. [ of water absorption ] According to the environment where the bridge formation object was placed, this shows that the volume changes and can also expect use to a mechano-chemical material.

[0020]

[Effect of the Invention] Even if it leaves it in a nature, it is decomposed by the microorganism which lives there, and the biodegradability quantity water absorption object of this invention is excellent in biodegradability.

[0021] Therefore, since it uses suitably as a soil conditioner by which after use is left in a nature, the coating agent of a seed, and a charge for vegetable cultivation of water retention agent material and the processing after Japanese lacquer and use becomes easy, the biodegradability quantity water absorption object of this invention can be used for sanitary items, the charge of disposable diaper material, etc., respectively.

[0022] Moreover, since the high water absorption object of this invention has the good compatibility to a living body, it can be used for the charge of artificial skin material again also at the charge of mechanochemical element material using the volume change by the perimeter environment.

[0023]

[Example] Next, an example explains this invention to a detail further.

[0024] The generation condition of the bridge formation object by change of one to example 12 quantity of radiation was examined. Pori (gamma-glutamic acid) produced by the microorganism was dissolved in water so that it might become concentration 5% of the weight, 0.5ml was poured distributively into each 1ml each vial bottle, and it covered. Exposure processing was carried out with each various quantity of radiation as shows a gamma ray in Table 1 with the gamma ray irradiation equipment which equipped each of these samples with the cobalt 60 (110TBq) as a line source. Each obtained sample processing object was picked out from each vial bottle, and a macromolecule and a decomposition product non-constructed a bridge were removed by carrying out a dipping to distilled water. In this way, each bridge formation object with high purity was acquired. About each [ these ] bridge formation object, the rate of gelation [the percentage of gel dry weight / the amount of brewing Pori (gamma-glutamic acid)] and water absorption (the moisture content / gel dry weight by which the maximum maintenance is carried out at the bridge formation object) were measured. The result is shown in Table 1.

[0025]

[Table 1]

	照射 量 M r a d	ゲル化率	吸水率
実施例1	1. 9 2	4 2	3 5 5 0
実施例2	3. 4 9	5 2	1 2 6 8
実施例3	5. 0 0	5 8	3 8 4
実施例4	8. 0 0	6 3	2 3 4
実施例5	10. 2 4	6 4	2 2 7
実施例6	12. 1 7	6 5	1 6 2
実施例7	14. 0 9	7 0	1 5 6
実施例8	16. 0 1	7 6	1 4 7
実施例9	19. 5 4	7 7	1 5 3
実施例10	23. 0 3	8 5	1 6 6
実施例11	25. 5 9	8 2	1 5 4
実施例12	29. 7 8	8 4	1 1 6

[0026] Although the bridge formation object formed by 1.92Mrad(s) had brittle quantity of radiation, water absorption is 3550, that is, about 3500 times as much water as the self-weight of a bridge formation object might be absorbed. Since the increasing inclination is seen, and the rate of gelation is large when water absorption has little quantity of radiation, and it decreases with the increment in quantity of radiation, and bridge formation is saturated with the increment in quantity of radiation when there is much quantity of radiation, it becomes almost the same, and it turns out anyway that water absorption exceeds 100 and sufficient absorptivity ability is shown.

[0027] The infrared absorption spectrum of the bridge formation object acquired in the example 5 which set quantity of radiation to 10.24Mrad(s) is shown in drawing 1. From this, it was checked that this bridge formation object is a bridge formation object of Pori (gamma-glutamic acid). Moreover, this bridge formation object was gel insoluble in water.

[0028] About the biodegradability of the acquired bridge formation object, it investigated as follows. That is, when each bridge formation object was buried and left in soil (field), it was decomposed until it was halved two months after, and, four months after, was decomposed completely.

[0029] The generation condition of the bridge formation object at the time of changing the concentration of the Pori (gamma-glutamic acid) water solution which performs an example 13 - 16 gamma irradiation was examined. Pori (gamma-glutamic acid) was dissolved so that it might become 2 - 5% of the weight, 0.5ml was poured distributively into each 1ml each vial bottle, and it covered. Exposure processing of the gamma ray was carried out with each quantity of radiation of 7Mrad(s) and 10Mrad(s), respectively with the gamma ray irradiation equipment which equipped each of these samples with the cobalt 60 (110TBq) as a line source. Each obtained sample processing object was picked out from each vial bottle, and a macromolecule and a decomposition product non-constructed a bridge were removed by carrying out a dipping to distilled water. In this way, each bridge formation object with high purity was acquired. Water absorption was measured about each [ these ] bridge formation object. This result is shown in Table 2.

[0030]

[Table 2]

	ポリマ濃度 (wt%)	吸水率	
		7Mrad	10Mrad
実施例13	2.0	693	580
実施例14	3.0	428	292
実施例15	4.0	348	254
実施例16	5.0	340	244

[0031] Although the gamma irradiation processing object was obtained like the example 13 except having changed the example Pori (gamma-glutamic acid) concentration of a comparison to 1% of the weight, as for this thing, neither generation of a bridge formation object nor gelation was seen.

[0032] It examined whether the bridge formation object of example 17 request could be repeatedly equal to use. The dipping of the bridge formation object of the example 16 generated by quantity-of-radiation 10Mrad was carried out to distilled water for one week, and it freeze-dried. The moisture content which holds at this time was measured, and water absorption was searched for. This was repeated several times. The result is shown in Table 3.

[0033]

[Table 3]

繰り返し回数	吸水率
1	244
2	248
3	256
4	266
5	281

[0034] Even if it uses a bridge formation object repeatedly, the property when using it first is held considerably, and this shows that it can be equal to repeat use.

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TECHNICAL FIELD

[Industrial Application] This invention relates to a new biodegradability quantity water absorption object and its manufacture approach. Since the biodegradability quantity water absorption object of this invention receives biodegradation in a nature, it can be used for the modifier in the engineering-works field, the soil conditioner in the plantation art field, a seed coating agent, the water retention agent for vegetable cultivation, the disposable diaper in makeup and the toiletries field, sanitary items, a mechano-chemical material, etc., for example.

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## PRIOR ART

[Description of the Prior Art] It is known by using the molecule bridge formation by repeating graft polymerization, radiation-induced crosslinking, ion bridge formation, bridge formation by the cross linking agent, freezing, and fusion to a water soluble polymer etc. that a high water absorption object will generate. These compounds are used abundantly after absorbing a water sorbent at the product which can be held so that it may not be leaked, for example, a disposable diaper, sanitary items, etc., using the property of unlike water absorbents, such as cloth and cotton, not repelling water even if it puts a pressure.

[0003] There are the starch system quantity water absorption object which carries out starch as such a water absorption object, carries out the graft polymerization of the synthetic monomers, such as an acrylic acid, to this as a raw material, and is acquired by forming three-dimension bridge formation, a hyaluronic acid system quantity water absorption object which is the water gel of a biological system, a polyvinyl alcohol system quantity water absorption object by which chemosynthesis was carried out, an acrylic system quantity water absorption object, an acrylamide system quantity water absorption object, etc.

[0004] These high water absorption objects will remain without being decomposed into the nature after use, when it is used in a nature like a soil conditioner. Moreover, it is very difficult to collect these. Then, development of the high water absorption object which has biodegradability which after use is decomposed also from environmental preservation by the microorganism which lives in a nature, and is incorporated by the carbon cycle cycle of a nature serves as pressing need.

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EFFECT OF THE INVENTION

[Effect of the Invention] Even if it leaves it in a nature, it is decomposed by the microorganism which lives there, and the biodegradability quantity water absorption object of this invention is excellent in biodegradability.

[0021] Therefore, since it uses suitably as a soil conditioner by which after use is left in a nature, the coating agent of a seed, and a charge for vegetable cultivation of water retention agent material and the processing after Japanese lacquer and use becomes easy, the biodegradability quantity water absorption object of this invention can be used for sanitary items, the charge of disposable diaper material, etc., respectively.

[0022] Moreover, since the high water absorption object of this invention has the good compatibility to a living body, it can be used for the charge of artificial skin material again also at the charge of mechanochemical element material using the volume change by the perimeter environment.

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TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] This invention conquers the fault which such a conventional high water absorption object has, and is made for the purpose of offering the high water absorption object which can aim at promotion of use in a nature and which has biodegradability.

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## MEANS

[Means for Solving the Problem] this invention persons came to make this invention for having the absorptivity in which the bridge formation object of the rate of predetermined gelation obtained by irradiating the radiation of predetermined quantity of radiation by the water solution or aquosity solution of predetermined concentration of Pori (gamma-glutamic acid) which is the water soluble polymer of biodegradability was excellent, without losing biodegradability based on a header and this knowledge, as a result of repeating research variously, in order to develop the high water absorption object of biodegradability.

[0007] That is, this invention offers the biodegradability quantity water absorption object which consists of the Pori (gamma-glutamic acid) radiation-induced crosslinking object of 40 - 90% of rates of gelation.

[0008] In this invention, the rate of gelation means the percentage of the dry weight of the Pori (gamma-glutamic acid) radiation-induced crosslinking object to the amount of preparation Pori (gamma-glutamic acid).

[0009] The biodegradability quantity water absorption object of this invention dissolves Pori (gamma-glutamic acid) in the mixed solvent of water or water, and a water-soluble solvent so that the concentration may become 2 - 5% of the weight preferably 1.5 to 6% of the weight, and after it irradiates a radiation at the solution subsequently obtained, it can manufacture it by separating the generated bridge formation object.

[0010] There is especially no limit about Pori (gamma-glutamic acid) used in this invention. Although based on what kind of manufacturing methods, such as a manufacturing method, a chemosynthesis method, etc. using a microorganism Thing [, for example, Biosci.Biotech.Biochem., produced by microorganisms, such as what produced by the microorganism, for example, a genus *Bacillus* kind like *Bacillus subtilis* etc., 56, 1031-1035 (1992), JP,1-174397,A reference] is desirable and what has 100 [ hundreds of thousands - ] and hundreds of thousands of molecular weight especially is desirable.

[0011] In this invention, although there is especially no limit about the water-soluble solvent used for dissolving Pori (gamma-glutamic acid) as a mixed solvent with water, it is good to make the rate of water into sufficient amount and to adjust the rate of a water-soluble solvent suitably so that Pori (gamma-glutamic acid) may be dissolved and it may get. As this water-soluble solvent, methyl alcohol, ethyl alcohol, an acetone, methyl acetate, ethyl acetate, etc. are mentioned, for example.

[0012] As for the solution which dissolved Pori (gamma-glutamic acid), it is desirable to put in, seal and use for a radiolucency container, for example, a glass vial bottle etc. Although there is especially no limit about the radiation irradiated by this solution, for example, alpha rays, beta rays, a gamma ray, an electron ray, a neutron beam, an X-ray, a charged-particle line, etc. are mentioned, it makes it possible to use a gamma ray preferably, and to obtain the specified substance by the shape of a film, since beta rays, an electron ray, and alpha rays have small penetrating power.

[0013] the case where a gamma ray is used for such radiation irradiation processing -- advantageous -- a gamma ray -- quantity of radiation, 1.0 to 50 Mrad, preferably, it irradiates by 2 - 30Mrad more preferably, and bridge formation usually advances at a room temperature 1.5 to

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[0017] The property of the biodegradability quantity water absorption object of this invention changes with the amount of radiation irradiation, or concentration of said solution, and a degree of cross linking serves as high water absorption small, and by the high exposure and high concentration, although bridge formation will be in a saturation state and serves as low water absorption relatively, it can hold absorptivity sufficient even in this case at a low exposure or low concentration.

[0018] The biodegradability quantity water absorption object of this invention can be used repeatedly. Even if it makes it dry by freeze-drying the bridge formation object which absorbed water and makes water swell again, it is changeless to absorptivity ability. Even if it repeated this several times, change was hardly seen by absorptivity ability.

[0019] Moreover, absorptivity ability changes with pH, and the biodegradability quantity water absorption object of this invention has low water absorption in acid pH field, and is high in alkaline pH field. [ of water absorption ] According to the environment where the bridge formation object was placed, this shows that the volume changes and can also expect use to a mechano-chemical material.

[0020]

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## EXAMPLE

[Example] Next, an example explains this invention to a detail further.

[0024] The generation condition of the bridge formation object by change of one to example 12 quantity of radiation was examined. Pori (gamma-glutamic acid) produced by the microorganism was dissolved in water so that it might become concentration 5% of the weight, 0.5ml was poured distributively into each 1ml each vial bottle, and it covered. Exposure processing was carried out with each various quantity of radiation as shows a gamma ray in Table 1 with the gamma ray irradiation equipment which equipped each of these samples with the cobalt 60 (110TBq) as a line source. Each obtained sample processing object was picked out from each vial bottle, and a macromolecule and a decomposition product non-constructed a bridge were removed by carrying out a dipping to distilled water. In this way, each bridge formation object with high purity was acquired. About each [ these ] bridge formation object, the rate of gelation [the percentage of gel dry weight / the amount of brewing Pori (gamma-glutamic acid)] and water absorption (the moisture content / gel dry weight by which the maximum maintenance is carried out at the bridge formation object) were measured. The result is shown in Table 1.

[0025]

[Table 1]

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実施例 3	5. 0 0	5 8	3 8 4
実施例 4	8. 0 0	6 3	2 3 4
実施例 5	1 0. 2 4	6 4	2 2 7
実施例 6	1 2. 1 7	6 5	1 6 2
実施例 7	1 4. 0 9	7 0	1 5 6
実施例 8	1 6. 0 1	7 6	1 4 7
実施例 9	1 9. 5 4	7 7	1 5 3
実施例 10	2 3. 0 3	8 5	1 6 6
実施例 11	2 5. 5 9	8 2	1 5 4
実施例 12	2 9. 7 8	8 4	1 1 6

[0026] Although the bridge formation object formed by 1.92Mrad(s) had brittle quantity of

radiation, water absorption is 3550, that is, about 3500 times as much water as the self-weight of a bridge formation object might be absorbed. Since the increasing inclination is seen, and the rate of gelation is large when water absorption has little quantity of radiation, and it decreases with the increment in quantity of radiation, and bridge formation is saturated with the increment in quantity of radiation when there is much quantity of radiation, it becomes almost the same, and it turns out anyway that water absorption exceeds 100 and sufficient absorptivity ability is shown.

[0027] The infrared absorption spectrum of the bridge formation object acquired in the example 5 which set quantity of radiation to 10.24Mrad(s) is shown in drawing 1. From this, it was checked that this bridge formation object is a bridge formation object of Pori (gamma-glutamic acid). Moreover, this bridge formation object was gel insoluble in water.

[0028] About the biodegradability of the acquired bridge formation object, it investigated as follows. That is, when each bridge formation object was buried and left in soil (field), it was decomposed until it was halved two months after, and, four months after, was decomposed completely.

[0029] The generation condition of the bridge formation object at the time of changing the concentration of the Pori (gamma-glutamic acid) water solution which performs an example 13 - 16 gamma irradiation was examined. Pori (gamma-glutamic acid) was dissolved so that it might become 2 - 5% of the weight, 0.5ml was poured distributively into each 1ml each vial bottle, and it covered. Exposure processing of the gamma ray was carried out with each quantity of radiation of 7Mrad(s) and 10Mrad(s), respectively with the gamma ray irradiation equipment which equipped each of these samples with the cobalt 60 (110TBq) as a line source. Each obtained sample processing object was picked out from each vial bottle, and a macromolecule and a decomposition product non-constructed a bridge were removed by carrying out a dipping to distilled water. In this way, each bridge formation object with high purity was acquired. Water absorption was measured about each [these] bridge formation object. This result is shown in

Table 2.

[0030]

[Table 2]

	ポリマー濃度 (wt%)	吸水率	
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[0031] Although the gamma irradiation processing object was obtained like the example 13 except having changed the example Pori (gamma-glutamic acid) concentration of a comparison to 1% of the weight, as for this thing, neither generation of a bridge formation object nor gelation was seen.

[0032] It examined whether the bridge formation object of example 17 request could be repeatedly equal to use. The dipping of the bridge formation object of the example 16 generated by quantity-of-radiation 10Mrad was carried out to distilled water for one week, and it freeze-dried. The moisture content which holds at this time was measured, and water absorption was searched for. This was repeated several times. The result is shown in Table 3.

[0033]

[Table 3]

繰り返し 回 数	吸水率
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2	248
3	256
4	266
5	281

[0034] Even if it uses a bridge formation object repeatedly, the property when using it first is held considerably, and this shows that it can be equal to repeat use.

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[Translation done.]

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- 2.\*\*\*\* shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

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DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] The infrared-absorption-spectrum Fig. of the bridge formation object generated by quantity-of-radiation 10.24Mrad of an example 5.

---

[Translation done.]

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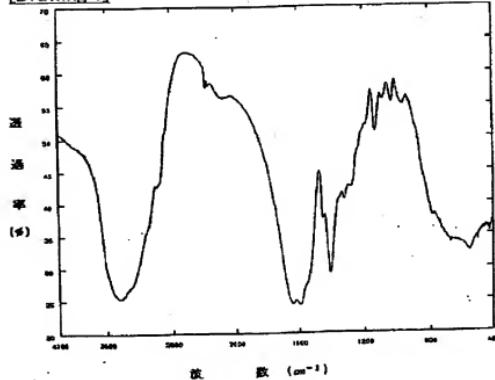
3. In the drawings, any words are not translated.

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DRAWINGS

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## [Drawing 1]



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[Translation done.]

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